

three-dimensional interconnected network. The three-dimensional interconnected network will usually be comprised of pores having diameters greater than about 20Å. The macrostructures can be made by forming an admixture containing a porous organic ion exchanger (e.g., a polymer-based ion exchange resin) and a synthesis mixture (e.g., for zeolite formation) capable of forming the porous inorganic material and the at least one metal; converting the synthesis mixture to the porous inorganic material; and removing the porous organic ion exchanger from the inorganic material. The metal-containing macrostructures find application in hydrocarbon conversion (e.g., hydrogenation, dehydrogenation, dehydrocyclization, isomerization, hydrocracking, dewaxing, reforming, conversion of alkyl aromatics, etc.) and in the reduction of emissions of hydrocarbons, carbon monoxide, and/or oxides of nitrogen from an internal combustion engine.

Please replace the paragraph at page 20, lines 1-21 with the following amended paragraph:

The organic ion exchangers suitable for preparing the macrostructures are organic porous materials with a surface charge and ion exchange capacity for anions or cations. Preferably, the organic ion exchangers are polymer-based which are sometimes referred to as ion exchange resins. Polymer-based ion exchangers are commercially available or can be readily prepared from resins that are commercially available. Examples of such resins include resins sold by Rohm and Haas Company under the registered trademark AMBERLYST (ion exchange resin) and resins sold by the Dow Chemical Company under the registered trademark DOWEX (ion exchange resin). These exchangers cover a broad spectrum of different cation and anion exchangers with varying ion exchange capacity, porosity, pore size and particle size. Ion exchangers with an apparent anion exchange capacity, typically greater than about 1 mEq/g of dry anion exchanger, are of special interest to the present invention. Macroreticular organic ion exchangers are particularly preferred in the practice of the present invention. By "macroreticular" as the term is commonly used in the resin art, it is generally meant that the pores, voids, or reticules are substantially within the range of about 200 to about 2,000 Å. Macroreticular resins are also referred to as macroporous resins.

Please replace the paragraph at page 41, lines 6-14 with the following amended paragraph:

A 1M HCl solution containing 0.01M palladium (II) chloride (PdCl<sub>2</sub>) was prepared in a beaker by dissolving the palladium

chloride in a 1M HCl solution. Next, a macroporous strongly basic anion exchanger sold under the tradename DOWEX MSA-1 [chloride form] was added to the contents of the beaker. The weight ratio of resin to Pd was 1:10 (10 grams of resin and 100 grams of Pd solution). Next, the beaker was placed in a shaker for one hour. The particles were then separated, rinsed several times with distilled water, filtered, and dried at 105°C.

Please replace the paragraph at page 45, lines 10-16 with the following amended paragraph:

DOWEX MSA-1 [chloride form] ion exchanger was exchanged with a 0.01M palladium (II) chloride ( $\text{PdCl}_2$ ) solution that was prepared by dissolving the palladium chloride in a 1M HCl solution. The weight ratio of resin to Pd was 1:10 (10 grams of resin and 100 grams of Pd solution). Next, the beaker was placed in a shaker for one hour. The particles were then separated, rinsed several times with distilled water, filtered, and dried at 105°C.

Please replace the paragraph at page 46, lines 11-15 with the following amended paragraph:

2 g of synthesis solution were mixed with 4 g of 0.1 M solutions of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ . Next, 2 grams of DOWEX MSA-1 [chloride form] ion exchanger was added to the mixture and the mixture was shaken for one hour. The particles were washed with distilled water and dried at 105°C for 2 hours.

A marked-up version of the amended paragraphs of the specification is attached hereto as ATTACHMENT A.

### *In The Claims*

Please amend claims 15, 16, 24, 34, and 36 as follows:

15. (Amended) The catalyst recited in Claim 4, wherein said at least one macrostructure contains less than 10% by weight of amorphous materials.

16. (Amended) A process of preparing a catalyst comprising; at least one porous macrostructure comprised of: (a) a three-dimensional network of self bound particles of porous inorganic material; and, (b) at least one metal, said particles occupying less than

75% of the total volume of said at least one macrostructure and being joined together to form a three-dimensional interconnected network comprised of pores; which method comprises the steps of: (i) forming an admixture comprising, a porous organic ion exchanger and a synthesis mixture which occupies at least a portion of the pore space of said porous organic ion exchanger and is capable of forming said porous inorganic material; (ii) converting said synthesis mixture to said porous inorganic material; and, (iii) removing said porous organic ion exchanger;

wherein said at least one metal is present in said admixture of step (i), or is added during the conversion of step (ii), or is added to at least one macrostructure before the removal in step (iii) of the porous organic ion exchanger and after the formation in step (ii) of the porous inorganic material, or is added to said at least one macrostructure after the removal in step (iii) of the porous organic ion exchanger, or combinations thereof.

24. (Amended) The process recited in Claim 20, wherein said porous polymer-based anionic ion-exchanger is a basic anion-exchange resin containing quaternary ammonium groups.

34. (Amended) The process recited in Claim 25, wherein said at least one macrostructure contains less than 10% by weight of amorphous materials.

36. (Amended) The process recited in Claim 25, wherein said particles are grown and joined together during conversion of said synthesis mixture to molecular sieve.

A marked-up version of the Amended Claims is attached hereto as ATTACHMENT B.

#### REMARKS

Claims 1-41 are pending. Claims 42-63 have been withdrawn from consideration pursuant to a restriction requirement. Various objections to the specification have been made. Claims 4, 5, 15, 19, 22, 24, 25, 34, and 36 are rejected under 35 USC § 112, 2 ¶. Claims 1-41 are provisionally rejected for obviousness-type double patenting over copending Application No. 09-315,869. Claims 1-4, 6-12, 14-24, 26-28, 33-36 and 38-41 are rejected under 35 USC 102(b) as anticipated by or, in the alternative, under 35 USC 103(a) as obvious over U. S. Patent

4,680,170 to Lowe et al (hereafter Lowe '170). Further, claims 1-41 are rejected under 35 USC 103(a) over Lowe '170 further in view of U. S. Patent 6,160,191 to Smith et al.

### **Election/Restriction**

Applicant affirms the election of claims 1-41 for examination.

### **Specification**

The abstract has been amended to add parenthetical examples or explanations of the terms "porous inorganic material," "metal," "porous organic ion exchanger," and "synthesis mixture." It has been further amended to set forth that the emissions are from a combustion engine and what the emissions are and to add parenthetical examples of hydrocarbon conversion processes. From the amended abstract one of skill would readily appreciate the nature of the catalyst being described without the need for and express example of a catalyst and what hydrocarbons are converted without need for examples thereof.

Substitute paragraphs have been submitted wherever a trademark appeared and such trademark has been capitalized and/or accompanied by the generic terminology.

### **Claim Objections**

Claim 16 has been amended to remove the comma between "step" and "(ii)."

### **Claim Rejections – 35 USC § 112**

Respecting claims 4 and 19 one skilled in the art would understand the metes and bounds of the "large" and "intermediate" sizes therein referred to. As the specification states at page 2, line 30 – page 3, line 2:

The pore size of crystalline microporous molecular sieves is usually in the range of from about 2 Å to **about 20 Å**.

At page 11, lines 15-18 the specification states that a **large** pore molecular sieve generally has a pore size of **at least about 7 Å** and includes LTL, VFI, MAZ, MEI, FAU, EMT, OFF, \*BEA, BOG, and MOR structure type zeolites (IUPAC Commission of Zeolite Nomenclature). At page 11, lines 22-26 the specification states that an **intermediate** pore size molecular sieves generally has a pore size **from about 5 Å to about 7 Å** and includes, for

example, MFI, MEL, MTW, EUO, MTT, MFS, AEL, AFO, HEU, FER, and TON structure type zeolites (IUPAC Commission of Zeolite Nomenclature). Accordingly, one skilled in the art would understand the porous inorganic material of claims 4 and 19 to be a molecular sieve having a pore size from about 5 Å to about 20 Å. Accordingly, claim 4 and 19 are not indefinite to one skilled in the art and applicant respectfully submits that this rejection should be withdrawn.

Respecting claims 5 and 25 which refer to “\*BEA,” one skilled in the art would understand that the asterisk in front of the code denotes a well-defined subunit for which pure end members have not been obtained. See [www.iza-sc.ethz.ch/IZA-SC/Atlas/data/BEA.html](http://www.iza-sc.ethz.ch/IZA-SC/Atlas/data/BEA.html) (print-out enclosed) for \*BEA which provides this explanation. Accordingly, claim 5 and 25 are not indefinite to one skilled in the art and applicant respectfully submits that this rejection should be withdrawn.

Respecting claim 22, the specification defines hydrothermal at page 25, lines 13-26 as follows:

Hydrothermal treatment refers to treatment in aqueous solution or aqueous suspension at a temperature exceeding 50°C, preferably exceeding 80°C and, in most cases, exceeding 95°C.

Accordingly, claim 22 is not indefinite to one skilled in the art and applicant respectfully submits that this rejection should be withdrawn.

Claim 24 has been amended to delete the term “strongly.” This moots the basis for rejection of claim 24.

Claim 34 has been amended to delete the term “significant” and to specify the limit of the amorphous material that the macrostructure may contain. Support for this amendment of claim 34 is found at page 11, lines 19-22 of the specification.

Claim 36 has been amended to delete the phrase “joined together by means other than by physical binding of the particles” and substitute therefore the phrase that the particles are “grown and joined together during conversion of said synthesis mixture to molecular sieve.” Support for

this amendment of claim 36 is found at page 23, lines 15-18 of the specification. This moots the basis for rejection of claim 36.

### **Applicants' Invention**

Applicant's invention is directed to metal-containing macrostructures of porous inorganic oxide, e.g., zeolite, and a method of producing the metal-containing macrostructures. One of the benefits of the metal-containing macrostructures is that they have a large interface between the gas or liquid phase of reactants and products and the solid phase of the porous inorganic material. For example, in the case of metal-containing macrostructures comprised of zeolite, the large interface allows good access of reactants to the channels of the zeolite. As a result, the metal-containing macrostructures can have enhanced performance when used in many commercial applications.

The macrostructures are made by: (a) forming a mixture of porous organic ion exchanger and a synthesis gel which is capable of forming said porous inorganic material; (b) converting the synthesis gel within the pore space of said porous organic ion exchanger to the porous inorganic material; and (c) removing the porous organic ion exchanger to obtain the macrostructures. The metal can be added to the macrostructures during their formation, after the macrostructures have been formed, or both.

The macrostructures include at least one metal and comprise a three-dimensional network of self bound particles of porous inorganic material. Preferably, the porous inorganic material comprises zeolite. The particles are joined together to form a three-dimensional interconnected network of pores and occupy less than 75% of the total volume of the macrostructures. Thus, more than 25% of the total volume of the macrostructures is unoccupied by the particles of porous inorganic material. The porosity of the three-dimensional interconnected network is external to the particles of the macrostructures.

Usually the porosity of the three-dimensional interconnected network of Applicant's macrostructures is not uniform and the size of the particles is usually small, e.g., less than 500 nm. The pores of the macrostructures provide openings for conducting reactants and products

between the exterior surface of the macrostructures and the inorganic oxide particles within the macrostructure.

Porous inorganic materials, e.g., zeolite, are used in a number of commercial applications, e.g., catalysts or catalyst supports for chemical processes.

Zeolites are crystalline microporous inorganic materials. One family of zeolite is aluminosilicate zeolite. Aluminosilicate zeolites are crystalline microporous inorganic materials that consist of a 3-dimensional structure of  $[\text{SiO}_4]^{4-}$  and  $[\text{AlO}_4]^{5-}$  coordination polyhedra linked by all of their corners. Within the zeolite structure are channels or pores which may be interconnected by a number of still smaller channels or pores. The dimensions of the channels are such as to allow adsorption of molecules with certain dimensions while rejecting those with larger dimensions.

Before using zeolite crystals in many of the commercial applications, it is usually necessary that they be shaped into macrostructures that are resistant to mechanical attrition, i.e., resistant to the formation of small particle fines, e.g., particles having a size of less than 20 microns.

For many applications, it is important that the macrostructures have a large interface between the gas or liquid phase of the reactants and products and the solid phase of the porous inorganic material of the macrostructures. The accessibility of the surface area of the macrostructure to reactant chemisorption and product desorption as well as the amount of surface area of macrostructures are significant factors in their effective use. Generally, macrostructures with a high surface area and having good accessibility to its surface area are more active than macrostructures without these characteristics. In the case of macrostructures comprised of zeolite, it is especially important that the macrostructures have a large interface so that the reactants have access to the channels/pores of the zeolite.

Many of the prior art techniques used to make macrostructures involve mixing zeolite with binding material to form an extrudable mass and then extruding the extrudable mass to form extrudates. Macrostructures prepared by this technique can have poor porosity because the binding material may block access to the pores of the zeolite and prevent hydrocarbons from

entering the pores of the zeolite and undergoing conversion within the channels. A further problem in the use of amorphous binder material is that it may itself catalyze undesirable reactions which can result in the formation of undesirable products. Still further, the binder material can dilute the active phase of the catalyst and thus affect activity of the catalyst.

### **Claim Rejections – 35 USC § 102/103**

Claims 1-4, 6-12, 14-24, 26-28, 33-36 and 38-41 are rejected under 35 USC 102(b) as anticipated by or, in the alternative, under 35 USC 103(a) as obvious over U. S. Patent 4,680,170 to Lowe et al (hereafter Lowe '170). Further, claims 1-41 are rejected under 35 USC 103(a) over Lowe '170 further in view of U. S. Patent 6,160,191 to Smith et al. These rejections stand or fall depending upon whether a *prima facie* case of obviousness of any claim over Lowe '170 has been properly established. Applicants respectfully submit that such a *prima facie* case of obviousness over Lowe '170 has not been established and request reconsideration in light of the remarks that follow.

The Examiner states that Lowe '170 Example 1 describes the same method for making a metal containing-zeolite catalyst as is used to make the catalyst of claim 1 here involved. With this the Applicant disagrees. Lowe '170 does not expressly teach (1) the use of a porous organic ion exchanger, (2) a synthesis mixture which occupies at least a portion of the pore space of said porous organic ion exchanger; (3) converting said synthesis mixture (which occupies at least a portion of the pore space) to said porous inorganic material; and, (4) removing said porous organic ion exchanger.

Because the Examiner viewed the production processes to be the same, the Examiner assumed that the products produced are the same and that these claims merely described the product of Lowe '170 by different terms than Lowe '170 used for the description of his product and that these different terms would have been obvious for one of skill in the art to have used to describe the product of Lowe '170. With this the Applicant disagrees. The zeolite product of Lowe '170 are crystals that are not (a) joined together to form at least one porous macrostructure; (b) a three-dimensional network of self bound particles; (c) particles occupying less than 75% of the total volume of said at least one macrostructure; and (d) joined together to form a three-dimensional interconnected network of pores.



To anticipate or render obvious the subject matter which claim 1 defines, it is the burden of the Examiner to establish by evidence that Lowe '170 describes or motivates one to produce a structure comprising

“(a) a three-dimensional network of self bound particles . . . occupying less than 75% of the total volume of said at least one macrostructure and being joined together to form a three-dimensional interconnected network of pores.”

Also, it is the burden of the Examiner to establish that the “particles” of the above described structure are themselves “of porous inorganic material” and further that the above described structure has “(b) at least one metal.”

Applicant respectfully submits that Lowe '170 fails to describe or to motivate one to produce a structure as above described. Lowe '170 does not describe or motivate the production of any structure of “self bound particles (which themselves are porous) . . . joined together to form a three-dimensional interconnected network of pores.”

Even if one assumes (as here the Examiner apparently did) that the “ion-exchange resin” which Lowe '170 utilizes is itself a “porous organic ion exchanger” (a fact which the Examiner has not established), nevertheless Lowe '170 does not produce a structure comprised of zeolite “self bound particles . . . joined together to form a three-dimensional interconnected network of pores.” The “100% ‘Silicalite’” which Example 1 of Lowe '170 produces are not “joined together to form a three-dimensional interconnected network of pores.” And the “BDH Dowex 1-XB(Cl) resin” which Lowe '170 uses in Example 1, even when “readily separated from the zeolite product” (Lowe '170, column 2, lines 56-58) does not comprise a structure of self bound zeolite particles joined together to form a three-dimensional interconnected network of pores.

Even if the “ion-exchange resin,” a/k/a the “BDH Dowex 1-XB(Cl) resin” of Example 1 of Lowe '170 is, or would be considered by one of ordinary skill in the art to be, a “porous organic ion-exchange resin,” such that, as the Examiner here hypothesizes, there might have been formed in the pores of such resin a “a three-dimensional network of self bound particles” of Silicalite this would be “a three-dimensional interconnected network of” Silicalite and not “of pores.” [Of course, the Examiner has not here established a *prima facie* showing by any evidence that, (1) the ion-exchange resin of Lowe '170 is a “porous” organic ion exchange resin,

or (2) within the ion-exchange resin of Lowe '170 there is formed a "three-dimensional network of self bound particles" of Silicalite, or (3) such Silicalite particles of Lowe '170 Example 1 are of a structure such that they are "occupying less than 75% of the total volume of" any macrostructure composed of particles "being joined together to form a three-dimensional interconnected network of pores."]

Lowe '170 does not direct one to a production of macrostructures. Lowe '170 directs one only to a production of particles of a zeolite. As Lowe '170 states:

The as-synthesised products, being free of alkali metal cations, have improved purity. The ion-exchange resin is readily separated from the zeolitic product and easily regenerated with cheap aqueous alkali metal hydroxide.

And as Example 4 of Lowe '170 shows the ion-exchange resin is regenerated and reused. This in and of itself suggest that the resin is not porous and/or that no zeolite forms within the ion-exchange resin, otherwise it would not be regenerable to its original state by a simple wash with an aqueous alkali metal hydroxide such that it could be reused.

The bottom line is the Lowe '170 does not remove the ion-exchange resin from anything in order to expose a three-dimensional network of self bound particles . . . being joined together to form a three-dimensional interconnected network of pores. A three-dimensional interconnected network of pores can not exist unless and until the ion-exchange resin is removed from something that first existed inside of the ion-exchange resin.

Lowe '170 does not describe the ion-exchange resin which it employees to be porous. Insofar as Lowe '170 says anything about the ion-exchange resin, Lowe '170 speaks only to the surface action of the ion-exchange resin, for example;

"The Applicants further believe that in some systems the surface of the resin may promote nucleation and reduce reaction times."  
(Col. 2, lines 26-28)

"Both ions could be introduced to the aqueous reaction mixture on the resin." (Col. 3, lines 15-16)

“In other cases the binding of impurity cations on to the resin will enable a purer zeolitic product to be obtained.” (Col. 3, lines 47-49)

“In the synthesis of some aluminosilicate zeolites, it is convenient to add at least some of the aluminium required to the reaction mixture in the cationic form bound to the resin. Cation exchange also provides a means of introducing unstable quaternary ammonium compounds to the reaction mixture. A quaternary bound to the resin is less likely to decompose than if it was in the solution phase and this is advantageous in the synthesis of zeolites which involve the use of such cations as the tetraethylammonium cation, for example zeolites beta, ZSM-12 and ZSM-20.” (Col. 3, lines 50-60)

If anything, Lowe ‘170 speaks of its ion-exchange resin as if it is not a porous resin. And Lowe ‘170 speaks to zeolite formation external of the resin, i.e., at the surface of the resin, not within the interior of the resin. Absent a porous resin one can not even begin to think of forming a three-dimensional network of self bound particles of a porous inorganic material (i.e., zeolite). And certainly Lowe ‘170 makes no statement whatsoever that relates to a three-dimensional network of self bound particles of a zeolite. Instead Lowe ‘170 describes its product zeolite only as crystals which are substantially uniform in size and composition (col. 2, lines 14-17). That is, Lowe’s zeolite crystals are free flowing, not self aggregated into a larger macrostructure. Lowe ‘170 does not speak to or suggest a macrostructure comprised of a network of self bound particles of a zeolite, much less does Lowe ‘170 speak to or suggest a macrostructure comprised of a three-dimensional network of self bound particles of a zeolite being joined together to form a three-dimensional interconnected network of pores.

Even if for the sake of argument one assumes that the ion-exchange resin of Lowe ‘170 were porous and that crystals of zeolite formed inside of the pores of the ion-exchange resin of Lowe ‘170 and formed a three-dimensional network of self bound particles of zeolite, this still would not be the subject matter of claim 1 here involved. Claim 1 specifies a porous

macrostructure, and this hypothetical structure of Lowe '170 would not be porous since Lowe '170 says nothing about separating the resin from the zeolite formed in the pores of the resin. All that Lowe '170 separates a resin from is from the zeolite crystals that form outside of the resin. So even if Lowe '170 formed a three-dimensional network of self bound particles of zeolite within the pore space of a resin Lowe '170 did not remove, nor does Lowe suggest the removal of, the resin from such a three-dimensional network of self bound particles of zeolite so as to leave a three-dimensional network of self bound particles of zeolite joined together to form a three-dimensional interconnected network of pores.

Lowe '170 does not teach or suggest the method of production or the product resulting there from as is here described and claimed. Accordingly, Lowe '170 neither anticipates nor render obvious any pending claim, nor does the combination of Lowe '170 with U. S. Patent 6,160,191 to Smith et al. render any claim to be obvious. Applicant respectfully submits that any rejection based on Lowe '170 should be withdrawn.

### **Double Patenting**

Claims 1-41 are provisionally rejected for double patenting of the obviousness type because claim 42 of commonly owned copending Application No. 09-315,869 reports that its mesoporous inorganic material may be silica, alumina or aluminum silicate. This alumina the Examiner says fulfills the "metal" requirement of the claims here pending. Applicant respectfully disagrees. The specification at page 8, line 24 – page 9, line 2 defines the "metal" of claims 1-41 as follows:

As used herein, the term "metal" refers to a metal that: (a) is present on the internal surface of the macrostructure or the external surface of the macrostructure, or both; and (b) affects the catalytic properties of the particles of the macrostructure or affects the adsorptive properties of the particles of the macrostructure, or both. The term "metal" does not encompass metal from which the porous inorganic material is comprised. For example, in the case of crystalline microporous molecular sieves, the term "metal" does not encompass any metal in the framework of the crystalline molecular sieve.

Hence the "metal" of pending claims 1-41 do not include the alumina of the mesoporous inorganic material of claim 42 of commonly owned copending Application No. 09-315,869. For

this reason double patenting of the obviousness type does not exist and the provisional rejection should be withdrawn.

### CONCLUSION

It is respectfully submitted that all issues and rejections have been adequately addressed and that all claims pending following entry of this Amendment are now allowable and that the case should be advanced to issuance.

If the Examiner has any questions or wishes to discuss the claims as amended, the Examiner is encouraged to call the undersigned at the telephone number indicated below.

Respectfully submitted,

September 26, 2002  
Date

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#### CERTIFICATE UNDER 37 CFR 1.8(a)

I hereby certify that this correspondence, and any attachments referred to, is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to Commissioner of Patents, Washington, D.C. 20231, on September 26, 2002.

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Date of Deposit

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Mia G. Marron



## ATTACHMENT A

### Marked-Up Version of Amended Paragraphs of the Specification

Page 59

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#### Abstract

#### METAL-CONTAINING MACROSTRUCTURES OF POROUS INORGANIC OXIDE, PREPARATION THEREOF AND USE

There is provided a catalyst containing porous macrostructures comprised of: (a) a three-dimensional network of particles of porous inorganic material (e.g., zeolites); and, (b) at least one metal (e.g., a catalytically active metal). The particles of the at least one macrostructure occupy less than 75% of the total volume of the at least one macrostructure and are jointed together to form a three-dimensional interconnected network. The three-dimensional interconnected network will usually be comprised of pores having diameters greater than about 20Å. The macrostructures can be made by forming an admixture containing a porous organic ion exchanger (e.g., a polymer-based ion exchange resin) and a synthesis mixture (e.g., for zeolite formation) capable of forming the porous inorganic material and the at least one metal; converting the synthesis mixture to the porous inorganic material; and removing the porous organic ion exchanger from the inorganic material. The metal-containing macrostructures find application in hydrocarbon conversion (e.g., hydrogenation, dehydrogenation, dehydrocyclization, isomerization, hydrocracking, dewaxing, reforming, conversion of alkyl aromatics, etc.) and in the reduction of emissions of hydrocarbons, carbon monoxide, and/or oxides of nitrogen from an internal combustion engine.

#### Page 20, lines 1-21

The organic ion exchangers suitable for preparing the macrostructures are organic porous materials with a surface charge and ion exchange capacity for anions or cations. Preferably, the organic ion exchangers are polymer-based which are sometimes referred to as ion exchange resins. Polymer-based ion exchangers are commercially available or can be readily prepared from resins that are commercially available. Examples of such resins include resins sold by

Rohm and Haas Company under the registered trademark [Amberlyst] AMBERLYST (ion exchange resin) and resins sold by the Dow Chemical Company under the registered trademark [Dowex] DOWEX (ion exchange resin). These exchangers cover a broad spectrum of different cation and anion exchangers with varying ion exchange capacity, porosity, pore size and particle size. Ion exchangers with an apparent anion exchange capacity, typically greater than about 1 mEq/g of dry anion exchanger, are of special interest to the present invention. Macroreticular organic ion exchangers are particularly preferred in the practice of the present invention. By "macroreticular" as the term is commonly used in the resin art, it is generally meant that the pores, voids, or reticules are substantially within the range of about 200 to about 2,000 Å. Macroreticular resins are also referred to as macroporous resins.

**Page 41, lines 6-14**

A 1M HCl solution containing 0.01M palladium (II) chloride ( $\text{PdCl}_2$ ) was prepared in a beaker by dissolving the palladium chloride in a 1M HCl solution. Next, a macroporous strongly basic anion exchanger sold under the tradename [Dowex] DOWEX MSA-1 [chloride form] was added to the contents of the beaker. The weight ratio of resin to Pd was 1:10 (10 grams of resin and 100 grams of Pd solution). Next, the beaker was placed in a shaker for one hour. The particles were then separated, rinsed several times with distilled water, filtered, and dried at 105°C.

**Page 45, lines 10-16**

[Dowex] DOWEX MSA-1 [chloride form] ion exchanger was exchanged with a 0.01M palladium (II) chloride ( $\text{PdCl}_2$ ) solution that was prepared by dissolving the palladium chloride in a 1M HCl solution. The weight ratio of resin to Pd was 1:10 (10 grams of resin and 100 grams of Pd solution). Next, the beaker was placed in a shaker for one hour. The particles were then separated, rinsed several times with distilled water, filtered, and dried at 105°C.

**Page 46, lines 11-15**

2 g of synthesis solution were mixed with 4 g of 0.1 M solutions of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ . Next, 2 grams of [Dowex] DOWEX MSA-1 [chloride form] ion exchanger was added to the

mixture and the mixture was shaken for one hour. The particles were washed with distilled water and dried at 105°C for 2 hours.

## **ATTACHMENT B**

### **Marked-Up Version of the Claims**

15. (Amended) The catalyst recited in Claim 4, wherein said at least one macrostructure [does not contain significant amounts] contains less than 10% by weight of amorphous materials.

16. (Amended) A process of preparing a catalyst comprising; at least one porous macrostructure comprised of: (a) a three-dimensional network of self bound particles of porous inorganic material; and, (b) at least one metal, said particles occupying less than 75% of the total volume of said at least one macrostructure and being joined together to form a three-dimensional interconnected network comprised of pores; which method comprises the steps of: (i) forming an admixture comprising, a porous organic ion exchanger and a synthesis mixture which occupies at least a portion of the pore space of said porous organic ion exchanger and is capable of forming said porous inorganic material; (ii) converting said synthesis mixture to said porous inorganic material; and, (iii) removing said porous organic ion exchanger;

wherein said at least one metal is present in said admixture of step (i), or is added during the conversion of step[, (ii)], or is added to at least one macrostructure before the removal in step (iii) of the porous organic ion exchanger and after the formation in step (ii) of the porous inorganic material, or is added to said at least one macrostructure after the removal in step (iii) of the porous organic ion exchanger, or combinations thereof.

24. (Amended) The process recited in Claim 20, wherein said porous polymer-based anionic ion-exchanger is a [strongly] basic anion-exchange resin containing quaternary ammonium groups.



34. (Amended) The process recited in Claim 25, wherein said at least one macrostructure [does not contain significant amounts] contains less than 10% by weight of amorphous materials.

36. (Amended) The process recited in Claim 25, wherein said particles are grown and joined together during conversion of said synthesis mixture to molecular sieve [joined together by means other than by physical binding of the particles].



## \*BEA

Type material:

Beta Na<sub>n</sub> [Al<sub>n</sub>Si<sub>64-n</sub>O<sub>128</sub>] with n 7  
 tetragonal, P4<sub>1</sub>22,  
 a=12.7, c=26.4 Å<sup>(1,2)</sup>

**Channels:** [001] 12 5.5 x 5.5\* <--> 100> 12 7.6 x 6.4\*\*

**Framework density:** 15.0 T/1000Å<sup>3</sup> (FD<sub>Si</sub>=15.3 T/1000Å<sup>3</sup>)

Coordinates of T-Atoms in the topological symmetry (P4<sub>1</sub>22)

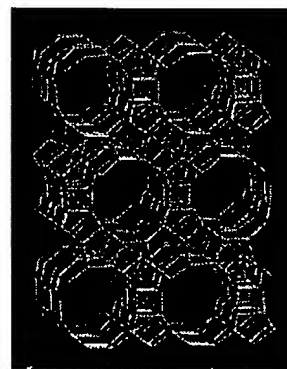
## Cell parameters:

a' = 12.632 b' = 12.632 c' = 26.186Å  
 alpha' = 90.000 beta' = 90.000 gamma' = 90.000°  
 volume' = 4178.15Å<sup>3</sup>

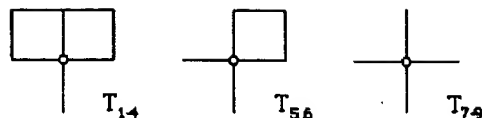
Atom	x	y	z	multiplicity	position
T <sub>1</sub>	0.9529	0.7960	0.0552	8	X, Y, Z
T <sub>2</sub>	0.9536	0.5429	0.0553	8	X, Y, Z
T <sub>3</sub>	0.7146	0.7921	0.0584	8	X, Y, Z
T <sub>4</sub>	0.7155	0.5451	0.0590	8	X, Y, Z
T <sub>5</sub>	0.3318	0.7884	0.0592	8	X, Y, Z
T <sub>6</sub>	0.3329	0.5409	0.0584	8	X, Y, Z
T <sub>7</sub>	0.5272	0.8594	0.1256	8	X, Y, Z
T <sub>8</sub>	0.5272	0.4728	0.1250	4	-Y+1, Y, +1/8
T <sub>9</sub>	0.1391	0.8609	0.1250	4	-Y+1, Y, +1/8

DLS R-value = 0.0022

Site Symmetry of T-positions: 8 T<sub>1-7</sub>(1) . 4 T<sub>8,9</sub>(2)



Loop configuration of T-atoms:



### Coordination sequences:

T <sub>1</sub>	(8)	4	10	19	32	51	77	105	133	167	207
T <sub>2</sub>	(8)	4	10	19	32	51	75	102	133	170	208
T <sub>3</sub>	(8)	4	10	21	32	49	76	109	137	170	207
T <sub>4</sub>	(8)	4	10	21	32	49	74	105	139	173	204
T <sub>5</sub>	(8)	4	11	18	29	48	80	107	133	160	203
T <sub>6</sub>	(8)	4	11	18	29	48	77	106	134	160	204
T <sub>7</sub>	(8)	4	12	18	31	51	76	109	133	164	210
T <sub>8</sub>	(4)	4	12	19	32	48	75	112	134	164	206
T <sub>9</sub>	(4)	4	12	17	30	54	77	106	134	160	212

### Materials with the same topology:

Borosilicate \*BEA<sup>(3,4)</sup>

Gallosilicate \*BEA<sup>(4)</sup>

Tschernichite<sup>(5)</sup>

### References:

1. J. M. Newsam, M. M. Treacy, W. T. Koetsier and C. B. DeGruyter, Proc. R. Soc. Lond. A **420**, 375 (1988).
2. J. B. Higgins, R. B. LaPierre, J. L. Schlenker, A. C. Rohrman, J. D. Wood, G. T. Kerr and W. J. Rohrbaugh, Zeolites **8**, 446 (1988).
3. M. Marler, R. Boehme and H. Gies, Proc. 9th IZC, Montreal, Butterworth-Heinemann (1993) p. 425.
4. K.S.N. Reddy, M.J. Eapen, P.N. Joshi, S.P. Mirajkar and V.P. Shiralkar, J. Incl. Phenom. Mol. Recogn. Chem. **20**, 197 (1994).
5. R.C. Boggs, D.G. Howard, J.V. Smith and G.L. Klein, Am. Mineral. **78**, 822 (1993).

NB: The asterisk in front of the code denotes a well-defined subunit for which pure end members have not been obtained. Further information can be found in (4). See also drawings of layer stackings

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